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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sensitivity of the radiative forcing by stratospheric sulfur geoengineering to the amount and strategy of the SO_2 injection studied with the LMDZ-S3A model" *by* Christoph Kleinschmitt et al.

Anonymous Referee #2

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This paper describes the use of the LMDZ-S3A model to study the efficacy and impacts of stratospheric sulfur geoengineering. This is valuable research, as stratospheric aerosols from many sources (e.g. volcanoes and hypothetical geoengineering schemes) can impact climate and chemistry, and there remains much uncertainty regarding stratospheric processes and aerosol evolution. This is an excellent model for this work, as the model includes aerosol microphysical processes, sectional aerosol size representation, aerosol-radiative interactions in both the SW and LW, high vertical resolution, stratospheric processes including QBO, and precursor gas emissions. The

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authors apply this model to study the impacts of equatorial SO2 injections at various magnitudes (2-50 Tg S/yr) and altitudes (15-23km) as well as a seasonal and broad injections, on aerosol properties, radiative fluxes, stratospheric circulation, and surface pollution. The paper is well-written, clearly laid out, and provides valuable contributions to scientific understanding of stratospheric sulfur geoengineering. I have one general/moderate concern and numerous small/specific concerns with the manuscript.

General/moderate concern:

The complexity of aerosol-chemical-dynamic processes, combined with long aerosol lifetime in the stratosphere, results in high uncertainty with models. Indeed, the results of this work differs fairly significantly from some other work, which the authors cite (for example, the large differences in radiative efficacy at higher injection rates compared to Niemeier and Timmreck 2015, and the large differences in efficacy at higher injection altitudes compared to Niemeier et al. 2011, Niemeier and Timmreck 2015). As such, I recommend a few things:

1) Add a paragraph to the introduction, as well as the conclusion, highlighting the significant uncertainties with stratospheric processes, especially hypothetical geoengineering schemes which have no observations to evaluate models against (yet) 2) Add more discussion throughout the paper on possible sources of uncertainties and errors with the current model implementation. A few discussion items: a) Prescribed oxidants: How do you know that prescribed oxidants in your model do not significantly impact results? It would be nice to do a sensitivity study, perhaps varying the availability of oxidants, and calculating how it impacts results. How might its feedback with water vapor impact results? Why did you not quantify ozone destruction, which is a known possible consequence of stratospheric sulfur geoengineering? Discuss. b) Coagulation processes: There are numerous coagulation processes that might be significant for geoengineered stratospheric aerosols – for example, you mention van der Waals forces. As you note, previous studies have found van der Waals forces to impact ambient stratospheric aerosol, and volcanic eruptions. The impacts of van der Waals forces

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on geoengineered aerosols seems uncertain, and possibly important. I suggest conducting an experiment with van der Waals forces compared to the standard experiment to see if it impacts results; at a minimum, further discussion. c) Any other limitations in your model worth discussing further?

Specific comments:

Abstract: 1) More clearly state that you calculate the limit of sulfate geoengineering efficacy to be 2 Wm-2, and that efficacy actually decreases at injections larger than 20 Tg S/yr – this is very interesting. 2) Add a caveat that stratospheric processes are complex and uncertain and that multiple modeling studies disagree with one another, including your model.

p1, line 20: suggest that you change the term "physico-chemistry" to something more understandable. Also, does this include impacts of ozone? If so, I suggest clarifying that, e.g. "chemistry and ozone"

p4, line 14: why do you expect the contribution of van der Waals forces to be insignificant? By changing the tails of the aerosol size distributions, van der Waals forces could possibly be significant over a long period of time, even if the impacts were negligable for a Pinatubo-size eruption. (see also my comment at p8, line 25).

p4, line 26: How might prescribed oxidants impact results or introduce error? (do you expect the oxidant concentration to remain constant over a long period of time given continuous so2 injection?)

p6, line 20: Change "completely" to something less exact, perhaps "essentially"

p8, line 25: The difference between your results and that of Niemeier and Timmreck (2015) is interesting. Can you estimate how much of the difference might be due to differences in the particle size distribution? (can you change your particle size distribution and run it through radiation code?) Other studies have found differences in the size distributions between modal models and sectional models. In an intercomparison

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study, Weisenstein et al. [2007] found sectional models to predict larger stratospheric aerosols after Pinatubo than modal models, and sectional models compared better to observations. Kokkola et al. [2009] completed an intercomparison between microphysical and sectional versions of the ECHAM model and found significant differences in the evolution of stratospheric aerosol size distributions, with differences between modal and sectional models increasing with increasing SO2 injection rate. English et al [2013] calculated aerosol mode widths from their sectional model assuming a log-normal distribution and found the mode width to change significantly after simulated volcanic eruptions, suggesting that 2-moment modal models with fixed mode widths may not accurately capture the evolving size distribution.

p9, line 32: Please cite Niemeier et al. 2011 who also studied injection height. The disagreement between your results and theirs is also interesting. Niemeier et al. 2011 also predicted increasing particle size with height (Fig 2b in their paper). Why might your results differ from theirs? Differences in LW radiation code? Differences in zonal transport with altitude? QBO?

p10, line 13: Are you injecting only in 28 grid boxes? If so, the concentrations might still be high compared to other studies which continuously inject across all grid boxes spanning a latitude range (e.g. 30 S - 30 N). Also, please clarify the altitude for the Broad injection here.

p11, line 15: Please cite Aquila et al. 2012, who also quantified the impacts that aerosol radiative heating have on the peak injection altitudes (for Mount Pinatubo).

p13, line 22: How does this contradict English et al. (2013)? They did not mention radiative fluxes. Differences in AOD distribution might be because their model does not have aerosol radiative heating coupled.

p13, line 22: Cite Niemeier et al 2011 in addition to Niemeier and Timmreck 2015. Again, why do your results differ if they also found an increase in particle radius with altitude?

p14, line 14: Expand this paragraph to further discuss the difficulties and complexities regarding geoengineered stratospheric aerosol (the processes are complex; the long lifetime allows for errors to amplifiy; geoengineering is only hypothetical and has no observations to evaluate models against (yet); potential uncertainties/errors in your model.

Figure 4: the x-axis units label appears as "m" which is incorrect.

Figure 6: latitude axis shows all "0"

Figure 7d, 8, 10d, 11,12, and 14: y-axis values are cut off

Figure 15: It would also be useful to have a plot of percent increase in deposition rate.

References

Aquila, V., L. D. Oman, R. S. Stolarski, P. R. Colarco, and P. A. Newman (2012), Dispersion of the volcanic sulfate cloud from a Mount Pinatubo–like eruption, J. Geophys. Res., 117, D06216, doi:10.1029/2011JD016968.

Kokkola, H., R. Hommel, J. Kazil, U. Niemeier, A.-I. Partanen, J. Feichter, and C. Timmreck (2009), Aerosol microphysics modules in the framework of the ECHAM5 climate model – intercomparison under stratospheric conditions, Geosci. Model Dev., 2, 97–112, doi:10.5194/gmd-2-97-2009.

Weisenstein, D. K., J. E. Penner, M. Herzog, and X. Liu (2007), Global 2-D intercomparison of sectional and modal aerosol modules, Atmos. Chem. Phys., 7, 2339–2355, doi:10.5194/acp-7-2339-2007.

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